[54]		IIDOGUANIDINE PLANT REGULATORS
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[52]		
[58]		ch
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[57] ABSTRACT

Novel compounds are disclosed which have utility as plant growth regulators. Use to produce growth regula-

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tors is specifically illustrated. The novel compounds of this invention are phthalimidoguanidines of the following general structural formulas:

$$\begin{array}{c|c}
O & R^{1} \\
\parallel & N-N-C=N-C=N-C
\end{array}$$

$$\begin{array}{c|c}
R^{2} & N-N-C=N-C=N-C
\end{array}$$

$$\begin{array}{c|c}
R^{4} & R^{3}
\end{array}$$

$$\begin{array}{c|c}
R^{4} & R^{3}
\end{array}$$

$$\begin{array}{c|c}
O & & \\
\parallel & & \\
N-N=C-N \\
\hline
 & & \\
N-N=C-N \\
\hline
 & & \\
R^2 & & \\
R^3 & & \\
\end{array}$$
(1b)

in which R, R¹ and R⁵ are hydrogen or C₁ to C₅ alkyl, R² and R³ are alike or unlike, one of which may be hydrogen but otherwise are substituents selected from C₁ to C₅ alkyl, alkenyl or alkynyl, branches or unbranched, C₂ to C₅ hydroxyalkyl or alkoxyalkyl, C₃ to C₅ cycloalkyl, furfuryl, pyridyl, benzyl, phenyl, C₁ to C₅ alkylphenyl, or —NR²R³ together may be 2,5-dimethyl-1-pyrryl, 4-morpholinyl or 1-pyrrolidinyl, R⁴ is halogen, nitro, cyano, trifluoromethyl, C₁ to C₅ alkyl or acyl and n is zero, one or two.

11 Claims, No Drawings

PHTHALIMIDOGUANIDINE PLANT GROWTH REGULATORS

DESCRIPTION OF THE INVENTION

This invention is directed to a novel group of chemical substances which have, to a high degree the ability to change or modify the growth habits of plants. The invention is also directed to methods of regulating growth of plants to obtain desirable modifications, in particular the increase of fruit set, by application to living plants, their seeds, or to the soil of effective amounts of the novel compounds. Also are included methods of manufacturing the compounds, as well as agricultural formulations containing the novel compounds as growth regulators.

Briefly, the novel compounds of this invention are phthalimidoguanidines of the following general structural formulas:

$$\begin{array}{c|c}
O \\
\parallel \\
N-N=C-N-C
\end{array}$$

$$\begin{array}{c|c}
R^5 \\
\downarrow \\
R^3
\end{array}$$

$$\begin{array}{c|c}
(1b) \\
(R^4)_n
\end{array}$$

in which R, R¹ and R⁵ are hydrogen or C₁ to C₅ alkyl, 35 R₂ and R³ are alike or unlike, one of which may be hydrogen but otherwise are substituents selected from C₁ to C₅ alkyl, alkenyl or alkynyl, branched or unbranched, C₂ to C₅ hydroxyalkyl or alkoxyalkyl, C₃ to C₅ cycloalkyl, furfuryl, pyridyl, benzyl, phenyl, C₁ to 40 C₅ alkylphenyl, or -NR²R³ together may be 2,5-dimethyl-1-pyrryl, 4-morpholinyl or 1-pyrrolidinyl, R⁴ is halogen, nitro, cyano, trifluoromethyl, C₁ to C₅ alkyl or acyl and n is zero, one or two.

Synthesis of the Growth Regulators

The compounds of this invention are readily prepared from the corresponding isothioureido isoindolediones by reaction with amines, as illustrated by the following procedure.

Preparation of 1,1,3-Trimethyl-3-phthalimido-2-phenylguanidine

$$\begin{array}{c|c}
O & CH_3 \\
N-N-C=N- \\
O & S-CH_3
\end{array}$$

$$\begin{array}{c|c}
ethanol \\
(CH_3)_2NH
\end{array}$$

$$\begin{array}{c|c}
O & CH_3 \\
N-N-C=N-\\
\longrightarrow O & N \\
CH_3 & CH_3
\end{array}$$

To a saturated solution of anhydrous dimethylamine in 80 ml. of ethanol was added 4.53 g. (0.01 mol) of 2-(1,2-dimethyl-3-phenylisothioureido)-1H-isoindole-1,3-(2H)-dione, and the resulting solution was heated to 50° C. for 30 minutes. Evaporation and treatment with heptane gave 3.5 g. of crystalline product, m.p. 146°-8° C.

The isothioureido isoindolediones employed as starting materials for the foregoing illustrative procedure may be manufactured by means of the general method comprising reacting a compound of the formula:

$$R \xrightarrow{Q} = Q \xrightarrow{R^1} H$$

$$Q \xrightarrow{N} Q \xrightarrow{N} Q$$

$$R \xrightarrow{N} Q \xrightarrow{N} Q$$

$$R^4)_n$$

in a non-reactive polar organic solvent with a compound of the formula R-X in which X is a leaving group exemplified by a chlorine, bromine, iodine or sulfate substituent. Specific procedures which are suitable for synthesis of representative compounds are outlined in the following chart and exemplified below.

At the first of the second second

$$N-H + H_2N-NH_2$$

$$N-NH_2$$

$$N-NH_2$$

$$N-NH_2$$

It will be understood that when it is desired that R and R¹ shall be different, the alkylation at the sulfur and amino locations is conveniently done in separate reaction steps. The following specific procedures illustrate the general methods of synthesis. All temperatures in the procedures are in °C.

Synthesis of methyl hydrogen phthalate

Methanol (300 ml) was added in a single portion to 148 g (1.00 mole) of phthalic anhydride and the resulting suspension was stirred and heated at reflux for 36 hours; solution occurred during heating. The solvent was removed and the product was recrystallized from a mixture of ethyl acetate and hexane to afford 114.6 g of the title compound (Lit.: Beilstein, 9,797-mp 82.5°, 84°).

Synthesis of methyl phthaloyl chloride

Methyl hydrogen phthalate (110.0 g., 0.611 mole) and thionyl chloride (77.4 g., 0.650 mole) were mixed in 200 ml of chloroform, keeping the temperature below 30°. After stirring for two hours at room temperature, the system was heated at reflux for five hours. The solvent was evaporated at reduced pressure and the crude product (120.9 g) was used without further purification (Lit.: Beilstein, 9, 797-no constants).

Synthesis of

2-(2-carboxybenzoyl)-1-methyl-N-phenylhydrázinethiocarboxamide (I)

This compound was made by reacting phthalic anhydride with 1-methyl-N-phenylhydrazinethiocarboxamide according to conventional procedures, as follows:

The hydrazinethiocarboxamide was dissolved in about 75 ml of dimethylformamide and placed in a 3-necked round-bottomed flask equipped with magnetic stirrer, condenser, additional powder funnel and thermometer. The anhydride was added in portions at 20°. The contents were stirred overnight at room temperature, then poured into ice water the following morning. The resulting solid was recrystallized in hexane and ethanol (m.p. 155°-156° C.).

Optionally ring substituted phthalic anhydrides and substituted 2-carbomethoxybenzoyl chlorides may be condensed with substituted 1-methylhydrazinethiocar-boxamides to give desired intermediate compounds as in the following scheme:

the late that the secont inued

or
$$+ H_2N - N - C - N$$
 CH_3
 $N - N - C - N$
 CH_3
 R^3
 CH_3
 R^4
 R^4

These methods are illustrated in the following specific procedures. The identity of the product was confirmed in each instance by means of infrared and nuclear magnetic resonance spectra. All melting points are as determined, uncorrected (Degrees C.).

N-Methyl-N-(phenylthiocarbamoyl)-2-amino-1H-isoin-dole-1,3-(2H)dione (II)

To an ice-cold solution of 8.25 g (0.025 mole) of 2-(2-carboxybenzoyl)-1-methyl-N-phenylhydrazinethiocar-boxamide in 225 ml of 1,2-dimethoxyethane at ~2° C., a solution of 5.5 g (0.027 mole) of N,N'-dicyclohexyl-carbodiimide was added dropwise below 5° C. with stirring. The mixture was stirred in the ice bath and then left at room temperature overnight. The mixture was filtered to remove N,N'-dicyclohexylurea and the filtrate was evaporated below 40° C., under vacuum, to give a yellow amorphous solid which was stirred in 100 ml of dry ether and warmed gently. The ether solution was allowed to stand for a few hours and filtered to give 4.6 g (59%) of whitish yellow crystals, m.p. 142°-144°.

Recrystallization from ethyl acetate-hexane gave 55 whitish crystals, m.p. 151°-153°.

Mass spectrum: M+311.

60

N-Methyl-N (phenylthiocarbamoyl)-2-amino-4-methyl-1H-isoindole-1,3-(2H)dione

To a solution of 6.8 g (0.037 mole) of 1-methyl-N-phenylhydrazinethiocarboxamide and 3.0 g of pyridine in 100 ml dry dimethoxyethane, 2-carbomethoxy-6-methylbenzoyl chloride (8.0 g, 0.037 mole) was added and the resulting mixture was stirred at room temperature for 60 hours. The solvent was distilled and the residue was taken up in ethyl acetate, filtered and dried on anhydrous magnesium sulfate. Removal of the sol-

vent gave 10 g (83%) of the desired product, m.p. 110°-115° (dec.).

2-Amino-1H-isoindole-1,3-(2H)dione

To an ice-cold suspension of 14.7 g (0.1 mole) of 5 phthalimide in 100 ml of 95% ethyl alcohol at 5° C., with stirring, 3.6 ml (0.11 mole) of 96.8% hydrazine was added dropwise. A slight exothermic reaction was observed and the mixture was allowed to stir at 5° C. for two hours. The mixture was diluted with 200 ml of ice 10 water, stirred, filtered, washed with water and dried to give 12.2 g (75%) of white powder, m.p. 199°-202°.

Recrystallization from methanol-water gave white needles, m.p. 201°-203°.

N-(Phenylthiocarbamoyl)-2-amino-1H-isoindole-1,3-(2H)dione (III)

To a suspension of 8.2 g (0.05 mole) of 2-amino-1Hisoindole-1,3-(2H)dione in 50 ml of dry 2-propanol, 6 ml (0.05 mole) of phenyl isothiocyanate was added. The 20 mixture was stirred and refluxed for 3 hours, allowed to cool to room temperature and poured into 300 ml of 50% ethyl alcohol. After stirring for one hour, the solid which formed was filtered, washed with water and dried to give 12.1 g (81%) of the desired product as a 25 white powder, m.p. 180°-181°. To make compounds in which Ar is benzoyl, benzoyl isothiocyanates may be employed in procedures of the type described above.

The free base was obtained by suspending the salt in dilute NH4OH and extracting with CHCl3 which was 30 washed with water, then with saturated aqueous NaCl and dried over anhydrous Na₂SO₄. Removal of the CHCl₃ at reduced pressure in a rotary evaporator gave

a residual oil which was crystallized from etherpetroleum ether, wt. 4.6 g, m.p. 90°-91° (75%).

Preparation of 2-(2-Methyl-3-phenylisothioureido)-1H-isoindole-1,3-(2H)-dione (IV)

To 100 ml of dry acetone, 5.0 g (0.018 m) of N-(phenylthiocarbamoyl)-2-amino-1-H-isoindole-1,3-(2H)dione and 2.5 g (0.018 m) powdered anhydrous potassium carbonate was added. To this suspension 1.1 ml (0.018 m) of methyl iodide was added and the mixture was stirred overnight at room temperature. The mixture was poured into ~ 300 ml of water, stirred, filtered, washed with water and dried to give 4.4 g 15 (78%) of a yellow powder, m.p. 161°-164°. Mass spectrum: M+311.

Preparation of 2-(1-Allyl-2-methyl-3-phenylisothioureido)-1-H-isoindole-1,3-(2H)dione

To 50 ml of dry acetone, 4.0 g (0.013 m) of the above product (IV), 2.2 g (0.016 m) of powdered anhydrous potassium carbonate, and 1.4 ml (0.016 m) of allyl bromide was added in succession and the mixture stirred overnight at room temperature. The mixture was poured into 300 ml of ice water with stirring. The aqueous layer was decanted and the residue suspended in fresh ice cold water, stirred and filtered to give a yellow semisolid product. This was purified by stirring in hexane to give a yellow powder 1.8 g (39%), m.p. 78°-81°.

Compounds which have been made by means of the foregoing illustrative procedures are listed in Table 1 below.

TADIE 1

					TABLE 1		
		· · · · · · · · · · · · · · · · · · ·	_(СОМРО	OUNDS OF THE FORMULAS): 	
		s to the second		\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\	$ \begin{array}{c c} R^{1} \\ N-N-C=N-\\ \end{array} $ $ \begin{array}{c c} R^{2} \\ \end{array} $ $ \begin{array}{c c} R^{2} \\ \end{array} $ $ \begin{array}{c c} R^{3} \\ \end{array} $	R^4) _n	(1a)
		s to the second			$\begin{bmatrix} 2 & N-N=C-N \\ 3 & N \end{bmatrix} = 0$ $\begin{bmatrix} N-N=C-N \\ N & N \end{bmatrix}$ $\begin{bmatrix} R^5 \\ N & N \end{bmatrix}$ $\begin{bmatrix} R^2 & R^3 \end{bmatrix}$	\mathbb{R}^4) _n	(1b)
		No. m.p. °C.	R	R ¹	$-NR^2R^3$	$(\mathbb{R}^4)_n$	R ⁵
	•	4368 oil	Н	CH ₃	$-N(C_2H_5)_2$	n = 0	
		4646 146-8°	"	"	$-N(CH_3)_2$	"	
		4647 192-5°	"		4-morpholinyl		
	•	4648 oil	"	11	-1-piperidyl	"	
	•	4649 159-61°	11	"	-1-pyrrolidinyl	"	w.w.e.
		4650 oil	11	**	—N—methyl benzyl	"	
		4651 oil	**	**	—N—isopropyl benzyl	"	<u></u>
	· · · .	4652 oil	**	"	—N—methyl phenyl	"	
		4653 oil	12	**	—N—ethyl phenyl	"	_
		4654 oil	**	"	$-N(CH_2CH_2CH_3)_2$	"	
•		4655 oil	**	"	CH ₃	"	
	• : .				$-N(CH-CH_3)_2$		
					11(011 0113)2		
		4656 oil	"	**	$-N(CH_2CH_2CH_2CH_3)_2$	**	
•		4657 oil	"	**	-N propyl phenyl	D	
		4658 oil	11	"	-N(sec.butyl) ₂	**	
		4659 oil .	"	"	-N(CH ₂ CH ₂ CH ₂ CH ₂ CH ₃) ₂	**	<u></u>
	\$ * .	4660 oil	**	"	$-N(allyl)_2$		
		4661 oil	"	**	−N(allyl)₂−N(nonyl)₂	11	
					· • /		

TABLE 1-continued

COMPOUNDS OF THE FORMULAS:

$$\begin{array}{c|c}
O & R^1 \\
\hline
1 & 2 & N-N-C=N-\\
\hline
5 & 3 & = O & N \\
R^2 & R^3
\end{array}$$
(1a)

O

$$1$$
 2
 $N-N=C-N$
 R^5
 R^5
 $R^4)_n$
 $R^4)_n$
 R^4

No.	m.p. °C.	R	R ¹	-NR ² R ³	$(\mathbb{R}^4)_n$	R ⁵
4702	oil	11	11	-N(isobutyl) ₂	**	
4703	oil	11	**	-N-methyl cyclooctyl	"	
4704	oil	11	"	$-N(\text{octyl})_2$	**	
4705	oil	11	**	-N-methyl propyl	"	_
4707	oil	"	**	-N-propyl isopropyl	"	
4708	oil	н	**	-N-propyl isobutyl	"	_
4709	oil	"	"	-N-propyl sec.butyl	"	
4710	oil	"	"	-N-propyl butyl	**	<u></u>
4716	oil		"	-N-benzyl 2-propynyl	"	
4717	oil	"	**	-N-allyl phenyl	"	
4718	oil	11	"	-N-ethyl p-tolyl	***	
4719	233-5°	11	"	$-N(CH_3)_2$	3-F	
4720	oil	"	"	-4-morpholinyl	"	
4721	190–191°	"	"	-NH-CH ₂ CH ₂ CH ₂ CH ₃	n = 0	
4722	136-8°	"	**	-NH-allyl	"	
4723	137-40°	"	**	-NH-benzyl	**	
4724	154-165°	"	**	-NH-(CH2)3CH3	3- F	
4725	52-3°	"	**	$-NH-C(CH_3)_3$	n = 0	_
4726	190-191°	"	**	-NH-CH ₂ CH ₂ CH ₃	"	
4727	185-95°	"	**	-NH-cyclopropyl	***	+
4728	oil	"	**	-NH-cyclopentyl	**	_
4739	52-4°	11	"	$-N(CH_3)(CH_2CH_2OH)$	•	
4740	glass	"	"	$-N(CH_3)_2$	$2,5-(CH_3)_2$	
4741	82-5°	"	**	$-N(CH_3)_2$	4-NO ₂	
4742	oil	"	**	$-N(CH_3)_2$	4-F	
4743	148-152°	"	**	$-N(CH_3)_2$	4-CN	" !!!-
4756	103-5°	"	"	$-N(CH_3)_2$	4-CF ₃	_
4757	137-9°	11	**	$-N(CH_3)_2$	2,4-Cl ₂	
4758	glass	11	11	$-N(CH_3)_2$	3-COCH ₃	
4759	glass	11		$-N(CH_3)_2$	n = 0	$-C_2H_5$
4760	glass	#		$-N(CH_3)_2$	n = 0	$-CH_3$
4762	164-72°	11	H	$-N(CH_3)_2$	3-F	
4764	oil	11	CH_3	1-pyrrolidinyl	11	
4850	oil	4-CH ₃	-	$-NH(CH_2)_3CH_3$	n = 0	
4956	oil	Η	"	-NH-CH ₂ CH ₂ OCH ₃	"	
4957	oil	"	21	-N-methyl furfuryl	**	
4958	oil	"	"	-2,5-dimethyl-l-pyrryl	**	<u></u>
4959	oil	"	**	-NH-2-pyridyl	"	····

Use of the Growth Regulators

In highly active compounds, phytotoxic and growthaltering effects of pre-emergent and post-emergent application are often readily apparent. These effects may be demonstrated by means of the following illustrative 55 procedures.

Pre-emergent Application

Disposable paper trays about 2 1.2 inches deep were filled with soil and sprayed with aqueous spray mixtures 60 1=slight effect, plants recovered, at a rate of 5 lb. of active chemical per acre of sprayed area, were seeded with 6 species of plant seeds and were then covered with about \(\frac{1}{4} \) inch of soil. The spray mixtures were prepared by dissolving the proper amount of growth regulant compound in 15 ml. of acetone, adding 65 4 ml. of a solvent-emulsifier consisting of 60 wt. percent of a commercial polyoxyethylated vegetable oil emulsifier (96 wt. percent active ingredient, Emulphor EL-

719), 20 wt. percent xylene and 20 wt. percent deodorized kerosene, then bringing total volume up to 60 ml. by addition of warm water. Twenty-one days after seeding and treatment the plantings were examined and plant injury was rated according to the following schedule.

DEGREE OF EFFECT

0 = no effect,

2=moderate effect, injury to 26 to 75 percent,

3=severe effect, injury to 76 to 99 percent, of foliage,

4=maximum effect (all plants died).

Post-emergent Application

Several species of plants were grown in potting soil in disposable styrofoam trays and tomatoes were grown in four-inch pots in the greenhouse. Aqueous spray formulations were prepared and the growing plants were sprayed at a spray volume of 60 gallons per acre and an application rate of 5 lb. per acre. Spray mixtures were prepared in the manner described above. For comparative purposes, plants were also sprayed at 60 gal./acre 5 with a spray mixture containing no growth regulant. Plant injury was again rated according to the schedule disclosed above.

Observations of growth regulant effects in both preand post-emergent tests were observed and recorded as 10 follows:

Effect	Abbreviation in Tables
Formative effect on new growth	F
Epinasty	E
Growth reduction	G
Necrosis	N
Non-emergence	K
Chlorosis	· C

In the table below there are tabulated the observations of pre- and post-emergent herbicidal and growth regulator effects resulting from use of one of the growth regulators of this invention according to the procedures set forth above. total volume up to 80 ml by addition of warm water. Of this spray mixture, a 50 ml portion was used to spray the plants at a rate of 3 lb. per acre of sprayed area. The remaining 30 ml. was diluted to 90 ml. with warm water and was used to spray the plants at a rate of 1 lb. per acre. One large, more mature tomato plants was included in the test along with the other, smaller growing plants. For comparative purposes, plants were also sprayed at a spray volume of 40 gallons per acre with a spray mixture containing no growth regulator.

Approximately fifteen days after spraying, the plants were observed and the results were evaluated according to the schedule disclosed above. Results obtained with representative compounds are presented in Table 2. The test species are so follows:

15 3. The test species are as follows:

	Number	Common Name	Scientific Name
	I	Pigweed	Amaranthus retroflexus
,	II	Lambsquarters	Chenopodium album
	III	Стаbgrass	Digitaria sanguinalis
	IV .	Downey brome	Bromus tectorum
	V	Giant foxtail	Setaria faberii
	VI	Nutsedge	Cyperus esculentus
	VII	Peanuts	Arachis hypogaea
)	VIII	Cotton	Gossypium herbaceum

TABLE 2

		-	<u>EFFE</u>	CTS OF	THE COM	<u> IPOUND</u>	S ON PI	LANT SPE	CIES			
		Pre	-emergen	t Effects					Post-e	mergent Eff	ects	
Compound No.	Digitaria sanguinalis	Celosia plumosa	Bromus inermis	Setaria italica	Raphanus sativus	Beta vulgaris	Setaria italica	Medicago sativa	Avena sativa	Raphanus sativus	Beta vulgaris	Lycopersicum esculentum
4368	F3G3	K4	F3G3	F3G3	F3G3	F3G3	FIGI	F3G2	F2G1	G1 N2F1	F3G2	F3G1

Post-emergent Application at Lower Rates on 24 Species

Twenty-four species of plants were grown in potting soil in disposable styrofoam trays and tomatoes were grown in four-inch pots in the greenhouse. Aqueous 40 spray formulations were prepared and the growing plants were sprayed at a spray volume of 40 gallons per acre and application rates of 3 lb. and 1 lb. per acre. The spray mixtures were prepared by dissolving the proper amount of growth regulator compound in 15 ml of 45 acetone, adding 4 ml of a solvent-emulsifier mixture consisting of 60 wt. percent of a commercial polyoxyethylated vegetable oil emulsifier (96 wt. percent active ingredient, Emulphor EL-719), 20 wt. percent xylene and 20 wt. percent deodorized kerosene, then bringing

IX	Tomato	Lycopersicum esculentum
X	Sugar beets	Beta vulgaris
XI	Wild buckwheat	Polygonum convolvulus
XII	Wild mustard	Brassica kaber
XIII	Mature tomato plant	Lycopersicum esculentum
XIV	Cocklebur	Xanthium pensylvanicum
XV	Morning glory	Ipomea purpurea
XVI	Soybeans	Soja max
XVII	Barnyard grass	Echinochloa crusgalli
XVIII	Green foxtail	Setaria viridis
XIX	Alfalfa	Medicago sativa
XX	Corn	Zea mays
XXI	Grain sorghum	Sorghum vulgare
XXII	Shattercane	Sorghum bicolor
XXIII	Wheat	Triticum aestivum
XXIV	Wild oats	Avena fatua
XXV	Rice	Oryza sativa.

TABLE 3

	POS	ST-EMER	RGENT EF	FECTS O	N 24 SPEC	IES	·					
	Appl'n. Rate											
Species	(lb/A)	4646	4677	4648	4649	4650	4651					
I	3	F2G2	F3G3	F3G3	F3G3	F3G3	F3G3					
	1	F1	F3G3	F3G3	F3G2	F3G3	F3G3					
II	3	F2G2	F3G3	F3G3	F3G3	F3G3	F3G3					
	1	F1	F2G2	F3G3	FIGI	F3G3	F3G3					
III	3	0	F1	F2G2	G1	F3G3	F2G1					
	1	0	0	F1G1	0	F1	F2G1					
IV	3	0	0	F1	0	FIG1	0					
	1	0	0	0	0	0	0					
V	3	0	Fl	F3G3	FÍ	F3G3	FIG1					
	1	0	0	F2G2	0	Fi	FIGI					
VI	3	0	0	0	0	F1	0					
	1	0	0	0	0	0	0					
VII	3	0 -	0	0	0	F1	FI					
	. 1	0	0	0	0	0	0					
VIII	3	0	F2	Fl	F2G2	F3G3	F3G1					

TABLE 3-continued

	PC			FECTS ON 24 SPE	CIES	·	-						
IX	1 3	0 F2	F1 G2F3	F1 F1 F3G3 F3G3	F2G1	F2G1 F3G3	·	• • • • • • • • • • • • • • • • • • • •	3	•			
v	1	FI	F3	F3G3 F3	F3G2	F3G2			:				
XI	1	F2 Fl	F1G1 F1	F3G3 F2G2 F3G3 F1	F3G3 F2G1	F3G3 F2G1	•						
	1	01 0 \ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \	· ·	1201	F2G2 F1	Fl	, ¹	τ.					
XII	1	0		F3G2 F1 F3G2 0					 - :	•		: ,,	
XIII	1	F1	F2 F2	F3 F2	F3	F3							•
	1	. 0		F2G1 F1	F2G2 F2G1	F2 F1							
XV	1	F1 0	F2 F1	F2G2 — 0	F3G3 F2G2	F3G2 F2G1		•				,	
XVI	3 1	F2 F1	G2F2 F2G1	F3G3 F2 F3G3 F1	F3G3 F3G3	F3G3 F3G2							
XVII	3 1	0	0	F1 0 0	FlG1 Fl	F1 0							
XVIII	3	0 0	F1 0	F2G1 F1 F1 0	F2G2 F1G1	F2G2 F1G1							
XIX		F1 0	F2 F1	F3G3 F2 F3G2 F1	F3G3 F3G2	F3G3 F3G1					· .		
XX	3	0	0	C1 0 0	Fl n	0							
XXI	. 3 1	0 0	F1	Fi 0	F2	F2 F1			3 -				
XXII	3	0	FI	F1 0	F2	F2							
XXIII	3	0	F1	0 0 0	F1	F1							
XXIV	3	0	0	0 0	F1	F1							
XXV	3	0	0	0 0	F1	F1							
Comments	Zoroninasta 16 € k	increased	0 increased	more more	more	more			•		•		
i te#i.		fruit set	fruit set	set set	set,	fruit set,	a sa					•	•
	Appl'n.			· · · · · · · · · · · · · · · · · · ·	tillers	tillers	•						
Species	Rate (lb/A)	***************************************	4653	Compound Nos. 4654 4655	4656	4657							
I	3	F3G3	F3G3	F3G3 F3G3	F3G3	F3G3	•	:			· ·		· ·
II	3	F3G3 F3G3	F3G3 F3G3	F3G3 F3G3 F3G3 F3G3	F3G3 F3G3	F3G3 F3G3				:	·		
III	3	F3G3 F3G2	F3G3 F3G2	F3G3 F3G2 F1 G1F1	F3G3 F2G1	F3G3 F1G1	· .						
ľV	3	F1G1 F1	F1 F1	F1 F1 0 0	FIG1 F1	F1 0			• .	1		. '	
V	3	0 F2	0 F3G2	0 F2G1 F2G1	0 F2G2	0 F2G2	ı						
VI	1 3	F2 0	F2G1 0	F2 F2G1 0 0	F1 0	F1 0				·	;		· • • • • • • • • • • • • • • • • • • •
VII		0 F 1	0 F1	0 0 F1 0	0 F1	0 F 1		•			•		•
VIII	1 3	0 F2G1	0 F2G1	0 0 F2 F1	0 F3G3	0 F2G1						:	
ΙX	1 3	F2 F3G3	F2 F3G3	F2 F1 F3G3 F3G3	F2 F3G3	F1 F3G3							•
X	. 1 3	F3G3.	F3G3 F3G2	F3G3 F3G2 F3G2 F3G3	F3G3 F3G3	F3G3 F3G3							
ΧI	3	F3G2 F2G2	F3G2 F2G2	F3G2 F3G2 F2G2 F2G2	F2G2 F2G2	F2G2 F2G2				•			
XII	3 **	F1G1 F2G2	F1 F3G2	F2 F1 F2G2 F3G2	F1G1 F2G2	Fl							
XIII	F 1.0	C2F1 F3G2	C2F1;	F2G1 C1F1 F3G3 F3G3	F1C2 F3G3	F1C1 F3G3							
ΚΙV	1 3	F3 F2G1	F3G2 F2G1	F3G1 F3G3 F2G1 F2G1	F3G2 F2G1	F3G3 F3G2	:						
: X V	1 3	F1 F3G2	F1		F1G1 F3G3	F1	:						
(VI	1 3 1	F3G2 F3G3	G2F2 F3G3	F3G3 F3G3 F3G3 F3G3 F3G2 F3G2	F3G3 F3G3 F3G2	F3G3 F2G1 F3G3							
(VII	3	F2G1	F2G1		. F1	F1	Ŧ						
XVIII .	1 3		,F1 ,F3G2	0 0 F2G2 F2G2	0 F2G2	0	47						
•		F2G2	F3G2	F2G2 F2G1 F3G2 F3G3	FIG1	F2G1							

F3G3 F3G3 F3G3 F3G3

XIX

TARIF 3-continued

	· · · · · · · · · · · · · · · · · · ·			TABLE 3	3-continu	ed			<u> </u>			Ţ.º Bm			
		POS	ST-EMEI	RGENT EF	FFECTS O	N 24 SPEC	IES					,			
	XX	. 3	F3G2 F1 0	F3G2 F2G1 F1	F3G1 F1 0	F3G1 F1	F3G2 F1G1	F3G1 F1	-			•			
	XXI	3	F3G1	F3G2	F2G1	Fi _.	F2G1	F2G1		:	· .				•
	XXII	3	F1 F3G1	F1 F3G2	F1 F2G1	F1 F1	0 F2G 1	F1 F2G1		·					
	XXIII	3	F1 F2G1	F1 F1G1	F1 F1	F1 0	F1	F1	;	. '	•				
	XXIV	3	FiGi	0 F1	0 F1	0	F1	0 F 1							
	XXV	3	0 F1G1	0 F1	F1	0	o F1	0 F 1							
	Comments	· 1	0 more fruit set	0 more tillers	0 more tillers	0	0 more tillers	0		i I					
		Appl'n.				-·····································		 	-						
	Species	Rate (lb/A)	4658	4659	Compo 4660	und Nos. 4661	4702	4703	_					•	•
	I	3	F3G3	F3G3	F3G3	F3G3	C3G3	F1 .							
	II	1 3	F3G3 F3G3	F3G3 F3G3	F3G2 F3G3	F3G3 F3G3	F3G2 C3G3	F1 F1	-		•	• •			· ·
•	III	3	F3G3 F2G2	F3G3 F1G1	F3G2 G2F1	F3G3 F3G2	F3G3 G2F2	F1 0							
	IV	1 3	FIGI FI	F1 0	G1 G1	F2G1 G1	F1 G1	0		• •					•
	V	1 3	0 F2G2	0 F1	0 F2G2	0 F3G2	0 F3G2	0 0					, -		
· .	VI	1 3	F1 0	0	F1 0	F2G1 0	F1 0	0							
	VII	1 3	0 F 1	0	0 F 1	0 F 1	0	0 0			¢	,			
	VIII	1 3	0 F2G1	0 F2G 1	0 F3G3	0 F2G1	0 F1	0 0					i .		• .
	IX	1	F1 F3G3	F1 F3G3	F1 F3G3	F1 F3G3	0 F3G3	0 F2			•				
	X	1	F3G3 F3G2	F3G3 F3G3	F3G3 F3G2	F3G3 F3G3	F3G1 F3G2	F2					· ř.		. '
	XI	1 3 .	F3G2 F2G2	F3G2 F2G1	F3G2 F1G1	F3G2 F1G1	F2 F1	F1 F1 0	:	e will be	: ·				
•	XII	1 3	F1G1 F2G2	F1 F2G1	F1 F3G2	F1: F3G3	0 C2F1	0 0							•
	XIII	3	F1C1 F3G3	F1 F3G3	G1F1 F3G3	F1G1 F3G3	0 F 3 G 1	0 F 3						,	
	XIV	1 3	F3G3 F2G2	F3G2 F2G1	F3 F2G1	F3 F2	F3 F1	F2 * F1							
	XV	1 3	F1 F3G3	F1 F3G3	F2 F3G2	F1 F3G3	F1 F3G2	F1 F2G2		· .			e ¹ *,		•
	XVI	1 3	F3G3 F3G3	F3G3 F3G3	F3G1 F3G3	F3G2 F3G3	F1 F3G2	F1 F3G2							
	XVII	1 3	F3G3 F1	F3G2 F1	F3G3 F1	F3G3 " F1	F3G2 0	F2G2 0	•		ofg t o an er	• .		•	
	XVIII	1 3	0 F2G2	0 F2G2	0 F2G1	0 F2G2	0 F2G1	0 0		<u>, 4</u> + 4 ·	•				
	XIX	1 3	F2G1 F3G3	F1 F3G3	F1G1 F3G2	F1 F3G3		0 F2C1	- † Beir			.1		•	
	XX	1 3	F3G2 F1	F3G1 F1	F3G2 F1	F3G1 F1	C1F2	F1 0				<u>.</u> :	<i>;</i>		
	XXI	1 3	F1 F2G1	0 F2	0 F2G1	0 F2G1	0 F2	0 F1						•	
		· 1	F2 F2G1	0 F2	F1 F2G1	F1 F2G1	F1 F2	0 F1	-				•		•
	XXIII	. 1 3	F2 F1	0 F1	F1 F1	F1 F1	Fi F1	0					-		
	XXIV	1 3	F1 F1	0 F1	0 F1	0	0 F 1	0 0	:						:
	XXV	1 3	0 F1	0 0	0 F1	0 F1	0 F1	0 n	· .		• •				· .
	Comments	1	0 more	0 more	0	0 more	0	0 more	•	•					
•	Comments		tillers	fruit	more fruit	fruit	more fruit	more fruit			:				· ·
			•	set	set, tillers	set, tillers	set, tillers	set	· · · · · ·						

	Appl'n.					.:		4,		1 1 3		_
	Rate			Co	mpound No	os.						•
Species	(lb/A)	4704	4705	4707	4708	4709	4710	-				
I	3	F3G3	F3G3	F3G3	F3G3	F3G3	F3G3	· · · · · · · · · · · · · · · · · · ·	. :		-	
	1	F3G2	F3G3	F3G3	F3G3	F3G3	F3G3	· . 1				

TABLE 3-continued

POST-EMERGENT EFFECTS ON 24 SPECIES											
II	3	F3G3	F3G3	F3G3	F3G3	F3G3	F3G3				
	1	F3G2	F3G3	F3G3	F3G3	F3G3	F3G3				
III	3	FI	F2G1	F3G2	F3G2	F3G2	F3G3				
177	I	0	0	FIG1	FiGi	FIGI	F1G1				
IV	3	G1	G1	G1	G1	G1.	G۱				
v	1 1	FIGI	F2G2	0 F2G2	F2G2	0 F3G2	F3G2				
• · · · · · · · · · · · · · · · · · · ·	1	n n	F1	F1	F2G2	F1	F2				
VI	3	ń	0	0	n 201	0	0				
* *	1	ñ	ñ	Û	n	0	0				
VII	3	Ô	Ö	Fl	n	Fi	Fl				
• • •	i	0	Ŏ.	0	0	Ô	0				
VIII	3	N3G2	Fl	F2G1	F2	F2	F2G1				
	1	NgF1	F1	F2	F1	F1	F1				
IX	3	N4	F3G3	F3G3	F3G3	F3G3	F3G3				
	1	F3G2	F3G2	F3G2	F3G2	F3G2	F3G2				
X	3	F3G3	F3G3	F3G3	F3G3	C3F3G3	F3G3C3				
	1 .	F2G2	F3G2	F3C2	F3G2C2	F2G2C2	F3G2C2				
XI	3	F1	Fi	Fl	Fl	F2G1	F2G1				
	1	0	0	0	0	0	F1				
XII	3	N4	C3F2	C3G3	C3G2	C3G3	C3G3				
	1	N2F2	FIC1	C2F1	C2F1	C2F1	C2F1				
XIII	3	N3F3	F3G2	F3G2	F3	F3C1	F3G1				
	1.	N2F2	F3	F3	F3	F3	F3				
XIV	3	FINI			F1	F1	· ·				
	1	Fl	F2	Fl	FI	Fi	FI				
XV	3	F3G3	F2G2	F3G3	F3G2	F3G3	F3G3				
	1	F2	F2G2	F2	F2G1	F2G1	F2G2				
XVI	.3	F3G3	F3G3	F3G3	F3G3	F3G3	F3G3				
	1	F3G2	F3G3	F3G3	F3G3	F3G3	F3G3				
XVII	3	0	FIG1	F2G1	F1	F2G1	F2G1				
	1	0	0	Ó	0	F1	Fi				
XVIII	3	F2G1	F2G2	F2G2	F3G2	F2G2	F2G2				
	1	Fl	F1	Fi	FI	F1G1	F!				
XIX	3	F3G3	F3G2	F3G3	F3G3	F3G3	F3G3				
	1	FINI	F3G1	F3G1	F2G1	F2G1	F3G2				
XX	-3	F1	F1	F1	F1	Fi	F2G1				
· ·	1.	0	0	F1	F1	0	F1				
XXI	3	F3G2	F2G1	F2G1	F2G1	F2G1	F3G2				
	1	F1	F1	F1	Fl	Fi	F2				
XXII	3	F3G2	F2G1	F2G1	F2G2	F2G1	F3G2				
	1	F1	Fl	F2	F2	Fl	F2				
XXIII	3	NIG1	F1	FI	Fl	FI	F1				
	1	0	0	F1	F1	0	0				
XXIV	3	NIGI	F1	. Fi	F1	Fl	F2				
·	1	0	F1	F1	F1	Fl	F1				
XXV	3	NIGI	F2 .	F1	F1	Fi	CIFI				
	1	0	0	F1	0	0	0				
Comments		more	more	more	more	more	more				
		fruit	fruit	fruit	fruit	fruit	fruit				
		set	set,	set,		set,	set,				
			tillers	tillers	tillers	tillers	tillers				
Aı	ppl'n.				······································						

	Appl'n.										•
	Rate	: ·	······································	Compou	ind Nos.				•	• .	·
Species	(lb/A)	4716	4717	4718	4719	4720	4721		٠.		
I	3	F3G3	F3G3	F3G3	F1	F3G3	F1				
	1	F3G3	F3G3	F3G3	0	F3G2	0			· · ·	
II	3	F3G3	F3G3	F3G3	Fl	F3G3	F1	•			
	1	F3G3	F3G2	F3G3	0	F3G3	0				
III	3	F2G1	F2G1	F1G1	0	F2G1	0				
	. 1	F1	F1	F1	0	FIGI	0			. •	•
IV	3	Fi	0	0	0	0	0				•
	1	0	0	0	0	0	0	• •			
V	3	F3G2	F3G2	F2G1	0	F2G2	0			•	
	1	F1	F1	FI	0	Fi	0				
VI	3	0	0	0	0	0	0				
	1	0	0	0	0	0	0				
VII	3	F1	F1	0	0	Fl	0			·	
	1	0	0	0	0	0	0				
VIII	3	F1	F2	Fi	0	F2	0				
	i	Fl	0	F1	0	F1	0			r	
IX	3	F3G3	F3G3	F3G3	Fl	F3G3	FI				
	1	F3G2	F3G1	F3G3	Fl	F3G2	Fl				
X	3	F3G3	F3G3	F3G3C2	F1	F3G3C3	Fl				
	1	F2G2C1	F2G1	F3C2	0	F2G2C1	F1	•			
XI	3	F1G1	F2G2	F2G1	0	FIGI	0				
	1	F1	Fl	F1	0	F1	0	e i e e e e e e e e e e e e e e e e e e			
XII	3	FlGI	C1F2G2	F2C2	0	F3C3G2	0				
	i	F1C1	FICI	F1C1	0	F2C2G1	0				
XIII	3	F3G1	F3G1	F3G1	FI		F1	i			5. 120 m

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TABLE 3-continued

- 	POS	т-еме	RGENT EF			CIES	
	1	F3	F3	F3	0	F2	0
XIV	3	F1	F2		0		0
XV	1.	F1 F2G2	F1 F3G3	F1 F3G2	0	F1 F3G2	0
7 k V	1	F2G1	F2G1	F2G2	0	F2G1	Ö
XVI	3	F3G3	F3G3	F3G3	0	F3G3	F 1
	l i	F3G3 F1	F3G3 F1	F3G3 F1	0 0	F3G3 F2G1	υ 0
	1	0	0	0	0	0	0
XVIII	3	FI	F2G1	F1G1	0	F3G2	0
XIX	1 3	F1 F3G2	Fi F3G1	F1 F3G2	O F1	F2G1 F3G3C1	o F1
74.74	1	F3	F2C1	F2C1	o ·	F3C1	0
XX	3	F1	F1	F1	0	FI	0
XXI	1 3	0 F2	F2	0 F2	0	F1 F2G1	0
7222	1	F1	Fl	F1	0	F1	0
XXII	3	F2	F2	F2	0	F2G1	0
XXIII	3	F1 0	F1 0	F1 0	·0	F1 F2G1	0
	1	0	0	0	0	0	0
XXIV	3	F1 F1	F1 F1	F! F!	0	F1 F1	0
XXV	3	Fl	F1 .	F2	0	FIGICI	0
	1	0	0	Fi	0	0	0
Comments		more fruit	more fruit	more fruit	more fruit	more fruit	more fruit
		set,	set,	set,	set	set,	set
 	······································	tillers	tillers	tillers	·	tillers	· · · · · · · · · · · · · · · · · · ·
	Appl'n. Rate			Compo	und Nos.	•	
Species	(lb/A)	4722	4723	4724	4725	4726	4727
I	3	F1	FlG1	F3G3	F3G3	0	F3G2
II	1 3	0 F1	0 0	F1 F3G3	F3G3 F3G3	0 0	G1 FIG1
,	1	Fl	0	F2G1	F3G3	Ö	0
III	3	0	0	0	F2G2	0	0
IV	3	0	0	.0	FIGI GI	.0	0
	1	0	0	0	0	0	0
V	3 1	0	0	0	F2G2 F1	0	0
VI	3	0	0	Õ	0	Ö	Ö
* ***	1	0	0	0	0	0	0
VII	3 1	0	0	0	0	0	0
VIII	3	0	0	Ö	F1	F1	N1
īV	1	0 C1E2	0 E2C2	0 E2C2	0 E2C2	0 E2C1	0 E2C3
IX	1	G1F2 F1	F2G2 F1	F3G3 F3G2	F3G3 F3G3	F2G1 F1	F3G3 F3G1
X	3	F2	F1	F3G3C1			F2G2
XI	. 1	F1 0	F1	F2G1 F1	F2G2C F2G2	1 0 0	F2
7 3.4 .	1	0	0	0	F1	0	Ŏ
XII	3	0	0	F1	F3G2C	_	F1
XIII	3	0 F3	0 F2	F3	F1C1 F3G3	0 F1	F3
XIV	1 3	F2 0	F2	F3 F1	F3G2 F2	F1	F2
222	1	0	0	0	F1	0	0
XV	3	F1	0	F2G1 F1	F2G2 F2G1	0	F1
XVI	3	F1	0	F2G2	F3G3	F1	N1F2
XVII	1	0	0 0	F2 0	F3G2 F1	0 0	N1 0
XVIII	1 2	0	0	0 F 1	0 E2C2	0	0
WA111	1	0	0	0	F3G2 F2G1	0	0
XIX	3	F1	F1 F1	F3G2	F3G3	F1	F2
XX	3	0	0 L1	F2 0	F3G2 F1	0.	F1 0
XXI	1 2	0	0	0 0	0 F2G 1	0	0
AAI	1	0	0	Õ	F2G1	0	0
XXII	3	0	0	0	F2G1	0	0.
XXIII	1 2	0 0	0 0	D ·	F2 0	0 n	0
2 x 2 x 4 1 1	1	0	0	ő	ő	ő	ŏ
XXIV	3	0	0	0	F1	0	0

TABLE 3-continued

	POS	ST-EME	RGENT E	FFECTS ON	V 24 SPE		······································							
XXV	3	0	0	0	0	0	0 -		•	•				
Commonto	i	0	0	0	0	()	0							
Comments	•	more fruit	more fruit		more fruit	more fruit								
		set	set		set	set								
	Appl'n.					·		, ",						
Species	Rate (lb/A)	4728	4739	Compos 4740	und Nos. 4741	4742	4743							
T	1	F3G3	F3G3	F3G2	F1G1	F3G3	F3G3							
•	1	G3F3	F3G3	F2G2	FIG1	F3G2	FlGI							
11	3 1	F3G3 F3G2	F3G3 F3G3	F3G2 F2B2	FIGI Fl	F3G3 F1G1	F3G3 F1G1							·
Ш	3	F2G2	F2G1	0	0	F2G2	0							
IV	1	FI F3G2	F1	0	0	0	0							
1 V	1	F2G2	0	0	0	0	0							
V	3	F2G2	F2G2	0	0	0	0							
VI	3	0	F1 C1	0	0	0	0							• .
	1	0	0	0	0	0	0							
VII	3 1	0 n	F1 0	0	0	0	0. 0	·						
VIII	3	0	F1	0	0	Ó	0							
IX	1	0 F3G3	0 F3G3	0 F2G1	0 G1F2	0 F3G3	0 F3G2							• •
171	1	F3G3	F3G3	FI	F2G1	F3G3	F2G1							
X	3	F3G3C2			F2	F3G2	Fi							
ΧI	3	F3G1 F1	F2G2 F1	Fl Fl	F1 0	F2G1 0	0 ~15.1		· · · · ·					. · ·
	1	0	F1	0	0	0	U						1 4	
XII	3 1	F2G2C1 F1C1	FIGI FICI	C1 0	0 ~ ~	F1 0	F1 0		-			•		
XIII	3	F3G3	F3G1	F3	F2	F3	F3							
XIV	1 3	F3G3 F2	F3 F1	F1	F1	F3	F2							
71.1 V	1	Fl	0	0	0	0	0							
XV	3	F3G3	F2G2	N2F1	N3F2	N3G3	N1							
XVI	3	F3G2 F3G3	F2G1 F3G3	F1	N1 F1	NIGI F3G3	0 F1						. ,	
	1	F3G3	F3G3	0	0	F2	0							
XVII	3 1	F1 0	F1 F1	0	0	0	0							
XVIII	3	F2G2	F2G2	0	0	Ö	0				•			
XIX	1	F1G1 F3G3	F1 F3G3	0 F 1	0 F 1	0 F3G1	0 C1F1							
«ኤሐ«ኤ	1	F3G3C2		F1	0	F2	C1F1							
XX	3	C1	F1	0	0	0	0							
XXI	3	F2G1	F2	0	0	. 0	0							•
373277	<u>l</u>	F1	F1	0	0	0	0			, .				
XXII	3 1	F2G1 F1	F2 F1	0	0	0	0							-
XXIII	3	0	G1	0	0	0	0							
XXIV	1 3	0 F1	0 F1	0 0	0 0	0 0	0 n							
	1	0	0	0	0	. 0	ŏ							
XXV	3 1	0 n	F1C1 F0	0 0	0 .	0	0	:						
Comments	•	more	more	more	тоге	niore	more	•			•			
		fruit set	fruit set	fruit set	fruit	fruit	fruit ·				•			•
	A no.12-	set	set	set	set	set	set						1	
	Appl'n. Rate	++		Compou	ind Nos.			_	. '					1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1
Species	(lb/A)	4756	4758 4	759 4760	4762	4764	4850							
I	3	F3G3	F1G1 0	0	F3G2	F3G3	F3G3							

	Appl'n.						·'	· ·i			•	
	Rate				Compo	ind Nos.			· · .			11 - 21 p
Species	(lb/A)	4756	4758	4759	4760	4762	4764	4850				
I	3	F3G3	FlG1	0	0	F3G2	F3G3	F3G3		•		
	1	F2G1	0	0	0	F2G1	F3G2	F3G3			•	
11	3	F3G3	F1G1	F1	0	F2G2	F3G3	F3G3	2. 1		:	•
	1	F2	0	0	0	F1	F3G3	F3G2				
Ш	3	0	0	0	0	0	FIGI	0				
	1	0	0	0	0	0	0	0	•			
IV	3	0	0	0	0	0	0	0	•			
	1	0	0	0	0	0	0	0	, in the second	1		
V	3	F1	0	0	0	0	F2G1	0	1			
	1	0	0	0	0	0	CÍ	0	• •			
VI	3	0	0	0	0	0	0	0		r		· · ·
	1	0	0	0	0	0	0	0	•			
VII	3	0	0	0	0	0	Fi	Ō		•		•
	1	Ō	0	Ö	0	0	F1	. 0		:		

TABLE 3-continued

	PO	OST-EME	ERGENT		ECTS ON		ECIES		· · · · · · · · · · · · · · · · · ·	· · · · · · · · · · · · · · · · · · ·	•	•	
VIII	3	F2	N1	0	0	F1	F2G1	N1		•			
	1	Fl	0	0	0	0	F2	0					
IX	3	F3G3	F2G2	0	0	Fi	F3G3	F3G3					
	1	F2	F2	0	0	F1	F3G2	F3					
X	3	F2	Fi	Fl	Fl	F1	F3G3C3	F2G1					
	1	F2	0	0	F1	F1	F2G2	F2					
XI	3	FIG1	0	0	0	0	FIC1	F1					
	1	0	0	0	0	0	Fl	0					
XII	3	F2	0	G1	0	0	F3G2C2	<u>·</u>					
	1	0	0	0	0	0	F1C1	FIGI					
XIII	3	F3	F3	NI	NIFI	F1	F3C2	F3G1					
	1	F2	F2	0	0	0	F3	F3					
XIV	3	F2	0	0	0	0	F2G1	Fl					
	1	0	0	0	0	0	F 1	F1					
XV	3	F2	N1	0	0	Fl	F3G2	F2G1					
	1	F1	0	0	0	0	F2G1	F2					
XVI	3 -	F2	NIFI	N1	F1	Fl	F3G3	F3G3					
	1	Fl	0	0	0	Fl	F3G3	F3G3					
XVII	3	F2G1	0	0	0	0	Fi	0					
	1	0	0	0	0	0	0	0					
XVIII	3	F2	0	0	0	0	F2G1	FIGI					
	I	0	0	0	0	0	F1	0					
XIX	3	F2	0	0	FI	F2	F3G2	F3G3	•				
	1	F1	0	0	0	F1	F2	F3					
XX	3	0	0	0	0	0	C 1	0					
	1	0	0	0	0	0	0	0					
IXX	3	F1	0	0	0	0	0	0					
4 ,4,4,4	1	0	0	0	0	0	0	0					
XXII	3	F!	0	0	0	0	0	0					
3/3/111	1	U	0	0	0	0	0	0					
XXIII	3	0	Ü	0	U	0	Fi	F1					
******	1	0	0	0	0	0	0	0		•			
XXIV	3	0	0	0	0	0	F2	F1		٠.		•	
VVV	I	0	U	0	0	U	0	0					
XXV	5	U	0	U	0	U	Fi	0		. •			
C	1	U	U	U	0	Ü	U	υ					
Comments					_ , , _ , _ ,		more tillers		_				• .
				App		<u></u>	mnound Nos						

		Appl'n.		~		
		Rate	·		ound No	
	Species	(lb/A)	4956	4957	4958	4959
	I	3	F3G3	F3G3	F3G3	F3G3
		1	F2G2	F3G3	F3G2	F3G3
	II	3	F3G3	F3G3	F3G3	F3G3
		1	F2G2	F3G3	F3G3	F3G3
	III	3	F1G1	F2G1	F2G2	F2G1
		1	0	0	G1F1	F1
	ΙV	3	G1	F1G1	F1G1	G1
		1	0	Ö	0	0
	V	3	F2G1	F2G2	F3G2	F2G2
•		i	0	FI	F2G1	F1
	VI	3	0	0	0	0
		1	0	0	0	0
	VII	3	Fl	Fl	F1	Fi
		1	0	0	Fl	0
	VIII	3	F2	F3G1	F3G2	F3G2
		1	F1	F1	F2	FI
	IX	3	F3G2	F3G3	F3G3	F3G3
		1	F3	F3G2	F3G3	F3G3
	X	3	F3G2	F3G2	F3G3	F3G3
		1	F2G1	F2G2	F3G2	F2G2
	· XI	3	F3G3	F3G3	F3G3	F3G2
		1	F2G1	FIG!	F2G2	F2G2
	XII	3	F2G1	F3G2	F3G3	F3G3
		1	F1	F2G1	F3G2	F2G2
	XIII	3	F3E3	F3G3	F3G3	F3G3
		1	F3#2	F3E1	F3E3	F3E2
	XIV	3	F1	F2	F2	F2
•	2 % A V	1	F1	F1	F2	F2
	XV	3	F2G2	F2G2	F3G2	F2G2
	# # T	1	F2G2	F2G2	F3G2	F2G2
	XVI	3	F3G3	F3G3		
	AVI	J 1			F3G3	F3G3
•	XVII	3	F3G2 F1G1	F3G3	F3G3	F3G3
	AVII	<i>3</i> 1	Fl	F!	F2	Fl E1
	XVIII	1		F1 F2G2	F1	F1
	V A 111	3	F2G1	F2G2	F3G2	F2G1
	VIV	. j	F1	Fl	F2	Fi
•	XIX	3	F3G2	F3G3	F3G3	F3G3
	XX	3	Fl	Fl	F2G1 F1	F2G2 F1
	ΛΛ	J	F 1	1,1	r i	I- 1

TABLE 3-continued

POST-EMERGENT E	FFEC	TS ON	24 SPECII	ES		
·	1	0	F1 F1	0		
XXI	3	F2	F2	F3G1	F2	
	i	Fi	F1	F2	F1	
XXII	3	F2	F2	F3G1	F2	
	1	F1	F1	F2	Fl	
XXIII	3	F1	Fl	F2G2	FIGI	
	1	0	F1	FIG1	0	•
XXIV	3	F1	F1	Fl	F1	
•	1	0	F1	F1	F1	
XXV	3	Fi	F1	F1	Fi	
	1	0	F1	Fl	0	

The use of many of the growth regulator compounds may be demonstrated by treatment of soybeans (Soja 15 max) to increase the number of seed pods and by treating tomato plants (Lycopersicum esculentum) to increase fruit set. In an illustrative experiment, Soja max (Evans variety) and Lycopersicum esculentum (Tiny Tim variety) were grown in 4-inch pots (one plant per pot) filled 20 with greenhouse potting soil (2 parts good top soil, $1\frac{1}{2}$ parts builders sand, $1\frac{1}{2}$ parts peat, fertilized with 5 lb. of 12-12-6 fertilizer and 5 lb. of finely ground limestone per cu. yd.). Aqueous spray formulations were prepared and the potted plants were sprayed at a spray volume of 25 40 gal. per acre and at application rates of 16, 4, 1 and $\frac{1}{4}$ oz. per acre. The spray mixtures were prepared by dissolving the proper amount of growth regulator compound in 15 ml. of acetone, adding 2 ml. of a solventemulsifier mixture consisting of 60 wt. percent of a 30 commercial polyoxyethylated vegetable oil emulsifier (96 wt. percent active ingredient, Emulphor El-719), 20 wt. percent xylene and 20 wt. percent deodorized kerosene, then bringing total volume up to 80 ml. by addition of a 0.156 wt. percent aqueous solution of liquid 35 non-ionic dispersant (90 wt. percent active trimethylnonyl polyethylene glycol ether, Tergitol TMN-10). Two replicates were sprayed at all application rates. For comparative purposes, plants were also sprayed at 40 gal./acre with water. The number of seed pods and of 40 fruit as percentage of arithmetic mean of the numbers on untreated plants was observed within approximately three weeks after spray treatment and the results are tabulated below. The extent of growth regulatory effect on the plants was estimated on a scale of 0 to 10 and is 45 also recorded in the following table:

TABLE 4

		TC	MATO	SOYBEAN		
Compound No.	Rate oz/A.	G.R. Effect 2 Rep. Avg.	Fruit Increase % of Check	G.R. Effect 2 Rep. Avg.	Pod Increase % of Check	
4368	16 4 1	6.5 4.5 2.5	150 236 192	7.5 4 3	203 155 121	

The information presented in tabular form herein will enable a worker in the art to make a selection from among the growth regulator compounds of the invention and to make some judgment with regard to application rates, depending upon the effect which is desired. It may be seen, for example, that total kills of some species of vegetation may occur at application rates as high as 5 to 10 lb. per acre, whereas beneficial effects may be observed on living plants at application rates of 1 lb. per acre or less.

The growth regulator compounds are usually applied in combination with inert carriers or diluents, as in aqueous sprays, granules and dust formulations in accordance with established practice in the art. An aqueous spray is usually prepared by mixing a wettable powder or emulsifiable concentrate formulation of a growth regulator with a relatively large amount of water to form a dispersion.

Wettable powders comprise intimate, finely divided mixtures of growth regulator compounds, inert solid carriers and surface active agents. The inert solid carrier is usually chosen from among the attapulgite clays, the kaolin clays, the montmorillonite clays, the diatomaceous earths, finely divided silica and purified silicates. Effective surfactants, which have wetting, penetrating and dispersing ability are usually present in a wettable powder formulation in proportions of from 0.5 to about 10 percent by weight. Among the surface active agents commonly used for this purpose are the sulfonated lignins, naphthalenesulfonates and condensed naphthalenesulfonates, alkylbenzenesulfonates, alkyl sulfates and non-ionic surfactants such as products of condensation of ethylene oxide with alkylphenols.

Emulsifiable concentrates of the growth regulator compounds comprise in each instance, a solution of growth regulator compound in a liquid carrier which is a mixture of water-immiscible solvent and surfactants, including emulsifiers. Useful solvents include aromatic hydrocarbon solvents such as the xylenes, alkylnaphthalenes, petroleum distillates, terpene solvents, etheralcohols and organic ester solvents. Suitable emulsifiers, dispersing and wetting agents may be selected from the same classes of products which are employed in formulating wettable powders.

In general, the growth regulators are applied in formulations which desirably contain from 0.1 percent to 95 percent of a compound of formula (1) and from 0.1 to 75 percent of a carrier or surfactant. However, direct application to plant seeds prior to planting may be accomplished in some instances by mixing powdered solid growth regulator with seed to obtain a substantially uniform coating which is very thin and comprises only one or two percent by weight or less, based on the weight of the seed. In most instances, however, a non-phytotoxic solvent, such as methanol is employed as a carrier to facilitate the uniform distribution of growth regulator on the surface of the seed.

When a compound is to be applied to the soil, as for a pre-emergence application, granular formulations are sometimes more convenient than sprays. A typical granular formation comprises the growth regulator compound dispersed on an inert carrier such as coarsely ground clay, or clay which has been converted to granules by treatment of a rolling bed of the powdered mate-

rial with a small amount of liquid in a granulating drum. In the usual process for preparing granular formulations, a solution of the active compound is sprayed on the granules while they are being agitated in a suitable mixing apparatus, after which the granules are dried 5 with a current of air during continued agitation.

I claim:

1. The method of regulating the growth of plants comprising applying to the plants, the seed or the soil an effective amount of a compound having one of the 10 following structural formulas;

in which R, R¹ and R⁵ are hydrogen or C₁ to C₅ alkyl, R² and R³ are alike or unlike, one of which may be hydrogen but otherwise are substituents selected from branched or unbranched C₁ to C₅ alkyl, alkenyl, alkynyl, C₂ to C₅ hydroxyalkyl or alkoxyalkyl, C₃ to C₅ cycloalkyl, furfuryl, pyridyl, benzyl, phenyl, C₁ to C₅ alkyl-substituted phenyl, or —NR²R³ together may be 2,5-dimethyl-1-pyrryl, 4-morpholinyl, or 1-pyrrolidinyl, R⁴ is halogen, nitro, cyano, trifluoromethyl or C₁ to C₅ alkyl and n is zero, one or two.

2. The method of regulating the growth of plants comprising applying to the plants, the seed or the soil an effective amount of a compound having one of the following structural formulas:

$$\begin{array}{c|c}
O & & \\
\downarrow & & \\
N-N=C-N \\
\hline
 & & \\
N-N=C-N \\
\hline
 & & \\
R^{2} & & \\
R^{3}
\end{array}$$
(1b)
$$\begin{array}{c}
(R^{4})_{n} \\
(R^{4})_{n}
\end{array}$$

in which R is H, R^1 is CH_3 , $-NR^2R^3$ is -1-piperidyl, R^5 55 is hydrogen or C_1 to C_5 alkyl and n is zero.

3. The method of regulating the growth of plants comprising applying to the plants, the seed or the soil an effective amount of a compound having one of the following structural formulas:

$$\begin{array}{c|c}
O & R^1 \\
\downarrow & N-N-C=N-C
\end{array}$$

$$\begin{array}{c|c}
\downarrow & N-N-C
\end{array}$$

$$\begin{array}{c|c}$$

in which R is H, R¹ is CH₃, —NR²R³ is —N(nonyl)₂, R⁵ is hydrogen or C₁ to C₅ alkyl and n is zero.

4. The method of regulating the growth of plants comprising applying to the plants, the seed or the soil an effective amount of a compound having one of the following structural formulas:

$$\begin{array}{c|c}
O & & & \\
\hline
 & & & \\
N-N=C-N \\
\hline
 & & & \\
R^{2} & & & \\
R^{3} & & & \\
\end{array}$$
(1b)
$$\begin{array}{c}
(R^{4})_{n} \\
R^{4} & & \\
\end{array}$$

in which R is H, R¹ is CH₃, —NR²R³ is —N-methylcy-clooctyl, R⁵ is hydrogen or C₁ to C₅ alkyl and n is zero.

5. The method of regulating the growth of plants comprising applying to the plants, the seed or the soil an effective amount of a compound having one of the following structural formulas:

$$\begin{array}{c|c}
O & & \\
\downarrow & & \\
N-N=C-N \\
\hline
 & & \\
& & \\
& & \\
R^{2} & & \\
\end{array}$$

$$\begin{array}{c|c}
R^{5} & & \\
& & \\
& & \\
& & \\
\end{array}$$

$$\begin{array}{c|c}
(1b) & & \\
& & \\
& & \\
\end{array}$$

$$\begin{array}{c|c}
(R^{4})_{n} & & \\
\end{array}$$

in which R is H, R^1 is CH_3 , $-NR^2R^3$ is $-N(\text{octyl})_2$, R^5 is hydrogen or C_1 to C_5 alkyl and n is zero.

- 6. The method of regulating the growth of plants which comprises applying to the plants pre- or post-emergently an effective amount of a composition comprising from 0.1 to 95 weight percent of a compound as specified in claim 1 in combination with a carrier or surface active agent.
- 7. The method of increasing fruit set of crop plants which comprises applying to the plant foliage an effective amount of a compound as specified in claim 1 in combination with an inert carrier and a surface active agent.
- 8. The method of regulating the growth of plants which comprises applying to the plants, a compound as specified in claim 1 in which the substituents represented by R, R¹,—NR²R³ and (R⁴)n are those tabulated below:

 $(\mathbb{R}^4)_n$

n = 0

n = 0

n = 0

n = 0

n = 0

n = 0

 $\mathbf{n} = \mathbf{o}$

n = 0

n = 0

n = 0

n = 0

n = 0

n = 0

n = 0

n = o

n = 0

n = 0

n = 0

n = 0

n = 0

n = o

n = o

n = 0

n = o

n = 0

n = 0

n = 0

n = 0

n = 0

4-nitro

4-fluoro

4-cyano

3-fluoro

4-CF₃

2,5-dimethyl

3-fluoro

 $-NR^2R^3$

 $-N(C_2H_5)_2$

 $-N(CH_3)_2$

-4-morpholinyl

-1-pyrrolidinyl

—N—methyl benzyl

—N—methyl phenyl

-N-ethyl phenyl

-N(isopropyl)₂

—N(sec. butyl)₂

 $--N(allyl)_2$

 $-N(isobutyl)_2$

—N—isopropyl benzyl

-N-(CH₂CH₂CH₃)₂

 $-N(CH_2CH_2CH_2CH_3)_2$

-N(CH₂CH₂CH₂CH₂CH₃)₂

-N-propyl phenyl

—N—methyl propyl

—N—propyl isopropyl

—N—propyl sec. butyl

-N-benzyl 2-propenyl

-NH-CH₂CH₂CH₂CH₃

 $-N(CH_3)$ (CH_2CH_2OH)

-N-propyl isobutyl

-N-propyl butyl

—N—allyl phenyl

-N-ethyl p-tolyl

-4-morpholinyl

 $-NH-C(CH_3)_3$

 $-N(CH_3)_2$

 $-N(CH_3)_2$

 $-N(CH_3)_2$

 $-N(CH_3)_2$

 $-N(CH_3)_2$

 $-N(CH_3)_2$

—NH—cyclopropyl

-NH-cyclopentyl

 R^1

H

H

H

H

H

H

H

H

H

Η

H

H

H

H

H

H

H

H

H

H

H

H

H

H

H

H

H

H

H

H

H

H

H

H

H

H

CH₃

CH₃

 CH_3

 CH_3

 CH_3

 CH_3

 CH_3

CH₃

CH₃

CH₃

 CH_3

CH₃

CH₃

CH₃

CH₃

 CH_3

 CH_3

 CH_3

 CH_3

 CH_3

 CH_3

 CH_3

CH₃

 CH_3

 CH_3

 CH_3

 CH_3

 CH_3

 CH_3

CH₃

CH₃

CH₃

 CH_3

CH₃

 CH_3

H

-continued

100

R	R ¹	$-NR^2R^3$	$(R^4)_n$
H	CH ₃	1-pyrrolidinyl	3-fluoro
4-CH ₃	CH_3	-NHCH ₂ CH ₂ CH ₂ CH ₃	n = o
H	CH_3	-NH-CH ₂ CH ₂ OCH ₃	n = o
H	CH ₃	-N-methyl furfuryl	n = o
H	CH_3	-2,5-dimethyl-1-pyrryl	n = o
H	CH_3	NH-2-pyridyl	n = 0.

9. The method of claim 7 in which the crop plants are of the species Lycopersicum esculentum.

10. The method of claim 7 in which the crop plants are of the species Soja max.

11. The method of regulating the growth of plants comprising applying to the plants, the seed or the soil an effective amount of a compound having one of the following structural formulas:

$$\begin{array}{c|c}
 & R^1 \\
 & R^1 \\
 & R^2 \\
 & R^3
\end{array}$$

$$\begin{array}{c|c}
 & R^4 \\
 & R^4 \\$$

$$\begin{array}{c|c}
O \\
N-N=C-N-C
\end{array}$$

$$\begin{array}{c|c}
R^5 \\
\downarrow \\
=O \\
R^2
\end{array}$$

$$\begin{array}{c|c}
(R^4)_n
\end{array}$$

in which R is hydrogen, R^1 is methyl, $-NR^2R^3$ is $-N(CH_3)_2$, $(R^4)_n$ is 3-acetyl, and R^5 is hydrogen or C_1 to C_5 alkyl.

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UNITED STATES PATENT OFFICE CERTIFICATE OF CORRECTION

Patent No	4,404,017	Dated September 13, 1983
Inventor(s)	Jerry L. Rutter	
It is and that sa	certified that error appoint and Letters Patent are he	pears in the above-identified patent ereby corrected as shown below:
	•	
Compound 4 Compound 4	•	line, "F3G2" should readF3G3; line, insertF3G2 in blank space;
•	le 3-continued, the firs 0 F1 F1 0	
		Bigned and Bealed this
		Third Day of December 1985
[SEAL]	Attest:	¬
		DONALD J. QUIGG
	Attesting Officer	Commissioner of Patents and Trademarks